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I. Radiation Transfer and Charge Particle Transport in Ionized Gases

Project Coordinator: A. C. Phelps

Plasma Spectroscopy (Dr. J. Cooper)

During the last few months we have made considerable progress, both theoretically and experimentally, in the understanding of the interaction of radiation with matter and in particular spectral line broadening.

For conciseness these advances will be listed. Firstly, the experimental program:

(1) Experiments on the Van der Waal's broadening of Cs and Si lines by Ar and Ne in a conventional pressure-driven shock tube have been completed.¹ These show the inadequacy of using a simple $1/r^6$ potential in the theory, since discrepancies of greater than two are found. Also, we expect to obtain high electron densities ($\sim 10^{18} \text{ cm}^{-3}$) at low temperatures (6000°K) for Stark broadening and other experiments by adding a cesium compound as an aerosol in this shock tube.

(2) Using a parallel-plate accelerator broadening by electrons of Ar II and Ca II lines has been measured.² These results, where comparison is possible, agree well with other investigations. However, for Ca II, H and K lines the disagreement with the precise quantum-mechanical calculations of Barnes and Peach³ is a factor of about two. Agreement is in fact better with semi-classical calculations⁴ using hyperbolic paths.

(3) The heat-pipe oven⁵ developed in collaboration with C. R. Vidal of the NBS Quantum Electronics Division is being used by a number of investigators for vacuum ultraviolet⁶ and other⁷ spectroscopic applications.

We have used it to measure collisionally induced fluorescence of the Li_2 molecule, and have designs to use the heat-pipe oven principle to make metal vapor lasers and other applications.

(4) A theta-pinch with an internal C-strap is being developed as a high temperature continuum light source. Effective temperatures of 2 μsec duration in excess of $50,000^\circ\text{K}$ have already been achieved.

The following are considered to be important advances in the theoretical program:

(1) Calculations of the broadening of hydrogen lines by electrons and ions using the "Unified theory" which we have developed⁹⁻¹¹ in collaboration with E. W. Smith and C. R. Vidal of the NBS Quantum Electronics Division give extremely good agreement between theory and experiment. Extensive tabulation of hydrogen Balmer and Lyman lines are now in preparation. The effect of time-ordering in these calculations has been shown to be relatively unimportant.¹⁴ The techniques of the Unified theory are now being used to investigate broadening due to neutral atom collisions, with specific emphasis on line wing behavior.

(2) Work in collaboration with E. W. Smith at NBS and Dr. W. Chappell of JILA relating the satellites seen on forbidden lines in helium to plasma turbulence has shown a direct correspondence between the profile and the turbulent spectrum.¹² This is very important for diagnosis of high temperature plasmas (the Russians in particular have a large effort in this field). More recently we have been able to relate the low frequency turbulence (specifically the autocorrelation function of the ion fields) to the actual shape of the forbidden line itself.¹³

(3) A quantum-mechanical theory of line broadening which takes into account the motion of the radiator and gives correlations between ordinary collision broadening and Doppler broadening (including "colli-

sional narrowing") has been developed^{15,16} in collaboration with E. W. Smith and others. Our results are essentially in agreement with those of Berman and Lamb,¹⁷ although their theory applies primarily to lasers and different techniques were used. We are investigating a semi-classical interpretation of this theory,¹⁸ and deviations from a Voigt profile of Van der Waals broadening under conditions of astrophysical interest are being considered. Using these techniques developed to handle Doppler effects, in conjunction with the Unified theory, it is hoped to obtain an improved formulation for the scattering of radiation under conditions when collisions are important. ,

(4) For the Stark broadening of isolated ion lines in the classical-path approximation rather complicated functions are required, since the trajectories are hyperbolic. These functions have been evaluated in collaboration with G. K. Oertel of NASA and a computer program is now available.⁴ Calculations of Stark broadening using this program are underway.² Similar classical-path functions for the broadening of neutral lines but with a Debye-shielded interaction potential have also to be evaluated.

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Scattering and Transport of Resonance Radiation (Dr. A. V. Phelps)

An essential step in the study of the scattering and transport of resonance radiation in gases is the accurate characterization of the profiles of the resonance lines. During the period of this report we have been assembling apparatus for use in measurements of the absorption profiles of the lines of the first resonance doublet of potassium. Thus far, we have assembled a vacuum system, filled an absorption cell with potassium, built an oven for use with the cell, and modified a cooled photomultiplier mount. We are currently realigning a monochromator. It is expected that most of the essential absorption profile

data for potassium will be obtained during the next report period. In addition, parts are being ordered for use in the construction of a dye laser. This laser will include the Lyot filter developed at JILA by Walther and Hall and will be used for measurements of the spectral distribution of the scattered light which results when the resonance state is excited with monochromatic radiation. It is expected that construction of this laser will be completed during the next period.

Radiation Transfer (Dr. D. G. Hummer)

Transfer in non-planar geometries. The major emphasis of work in this project has been two-fold: (1) to develop practical computational methods for the solution of radiative transfer equations under non-LTE conditions in geometrical configurations other than plane-parallel slabs, and (2) to recognize and understand the basic features of the radiation field in such geometrical configurations which are characteristic of that particular geometry.

The simplest, and in many ways most important, non-plane geometrical configuration is the spherically-symmetric system. Although such systems have been studied for many years, prior to the work of Chapman¹ an important feature of these problems was unnoticed, namely that the radiation field became sharply outward peaked for large radii. Because this basic property was not accounted for, the asymptotic results first obtained by Chandrasekhar² as well as many subsequent calculations are qualitatively as well as quantitatively wrong.

Under this project, Hummer and Rybicki developed an efficient procedure for solving this class of problem for an arbitrary dependence

of opacity with radius. The basic idea is to retain only the first two moments of the transfer equation, which involve the first three moments of the intensity, namely $J(r)$, $F(r)$ and $K(r)$. Then if some closure conditions say that $f(r) = K(r)/J(r)$ is assumed to be known at each point, then the moment equation can be solved if a boundary condition is available. The familiar Eddington approximation is one such choice ($f=1/3$) but this completely fails to account for the outward peaking of radiation field mentioned above. Recently, in the defense industry, calculations have been performed using an approximate a priori specified function $f(r)$. Hummer and Rybicki have determined $f(r)$, which is called variable Eddington factor, iteratively, by alternating between the moment equations and the formal solution evaluated by an extension of the procedure due to Featrier,³ using a coordinate system capable of representing the outward-peaking in a non-singular way. This non-linear iteration scheme was found to converge uniformly and very rapidly, so that three iterations gave an accuracy of better than 1%. The asymptotic behavior of the radiation field was discussed in considerable detail, as were some useful approximate solutions. An account of this work has been submitted for publication in the Monthly Notices of the Royal Astronomical Society.

Subsequently, Cassinelli and Hummer have generalized the method of Hummer and Rybicki to treat non-conservative problems and linearly polarized radiation. This work shows that radiation emitted at large angles to the normal is very strongly polarized as a consequence of the strong outward-peaking of the radiation. An account of this work is being prepared for publication in Monthly Notices.

It appears that the general technique of iterational determination of a closure condition for moment equations should be valuable for other problems, including transfer problems in general three-dimensional geometries. Investigation of these possibilities is high on our list of priorities.

General frequency redistribution in spectral line formation. For an accurate treatment of the transfer of line radiation in a low density gas, it is necessary to solve the transfer equation taking into account the changes in frequency of a photon as it is scattered. The relation of the incident and final frequencies and directions is specified by the so-called redistribution function; the specification of these functions for particular broadening mechanisms has been discussed by Hummer.⁴

Because of our interest in developing a model for transfer in a turbulent "clumpy" medium, in which one first determines the spectral response of a clump or cloud to an incident photon given frequency, and then considers a gas of such clouds, thus minimizing the behavior of the atoms in a real gas, it became essential to be able to perform calculations of the type described in the preceding paragraph in an efficient manner. Some solutions had been obtained earlier by Hummer,⁵ but a more general approach was indicated. To this end, Adams, Hummer and Rybicki developed a very fast generating procedure for the redistribution function appropriate for combined natural and Doppler broadening (the so-called R_{II} function). They further developed a representation of the frequency dependence of radiation field in terms of the so-called "spline functions" that was capable of efficiently handling the very large band widths characteristic of problems of this type.

This novel approach should be very useful in a wide variety of other transfer calculations. An account of this work is now in press in the Journal of Quantitative Spectroscopy and Radiative Transfer.

Dr. Adams has prepared a package of computer programs to generalize the coefficients needed in the spline function representation for the function R_{II} and to solve the transfer problem for this situation in a plane-parallel geometry in which all atmospheric parameters can be arbitrary functions of depth. This program has been used so far primarily to study the penetration of an external field into a slab and to investigate the theoretically interesting question of the mean number of scatterings experienced by a typical photon. The application to the cloud model has had to be shelved temporarily for lack of support.

Radiative transfer with the migration of excited atoms. The recent experimental investigations by Phelps and Chen⁶ on the transfer of the first resonance line in cesium vapor have indicated the importance of the migration of excited atoms as an energy transfer mechanism competitive with and closely related to radiative transfer. An analysis of this and similar experiments offers the opportunity of inferring momentum transfer cross sections between excited states and ground state atoms. An analysis by Hummer of this problem produced a computationally exact solution of the problem in which the transfer of energy by radiation and by the motion of the excited atoms was treated simultaneously and on the same footing. Mr. Kunasz has prepared a very flexible computer program based on this analysis and its application to the interpretation of experimental data will begin shortly.

Basic mathematical functions of spectral line formation. For many laboratory and engineering applications involving the transfer of resonance radiation, five basic functions defined in terms of integrals over the absorption coefficient and exponential and exponential-integral functions are necessary. The evaluation and tabulation of these functions in a systematic and practical way continues as a low-priority background project. A pilot calculation for the Lorentz profile has been successfully completed (at very little cost) and tables made available privately to interested users. It is planned to extend this work to include a number of Voigt profiles before publishing all of these functions in a convenient tabulation.

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Plasma Statistics (Dr. W. E. Brittin and Dr. W. R. Chappell)

Dr. Chappell has continued his work on weakly ionized plasmas. During this period a paper on electron diffusion in weakly ionized cases was written in collaboration with Dr. Williams of N.O.A.A. As in the unmagnetized case a correction to Weinstock's¹ work was obtained arising from collective interactions. The previous paper on the unmagnetized case has been accepted for publication by Physics of Fluids and will appear in the near future. Experimental results obtained in the last few months by workers at N.O.A.A.² show a remarkable agreement with our theory and indicate that the measurement of the density autocorrelation function by means of two electrostatic probes may provide a sensitive diagnostic tool. The paper on the magnetized case has been submitted to Physics of Fluids. Experiments relating to the magnetized case will be begun at N.O.A.A. in the near future.

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II. Ionization Kinetic and Reaction Rates

Project Coordinator: Dr. W. C. Lineberger

Ionization Kinetics (Dr. W. C. Lineberger and Mr. T. A. Patterson)

The initial results of the near threshold S^- photodetachment measurements have been published in Physical Review Letters.¹ Our rather qualitative coupling scheme required to explain the relative strengths of the various fine structure transitions has been confirmed by much more elaborate calculations of Rau and Fano at the University of Chicago. Their calculations also indicate the possibility of finding resonances in the photodetachment cross section resulting from electron-atom interactions. This possibility will be pursued in a series of high resolution measurements using the Lyot filter developed by Walther and Hall² at JILA. To accomplish this task, we have constructed a new, better engineered flashlamp-pumped dye laser to be utilized with a Lyot filter. In order to make use of this high resolution, it has also become necessary to significantly automate our previously rather crude data acquisition system. These systems are substantially completed, and are currently in the process of being checked out. We anticipate significant results during the next six months.

The nitrogen laser-pumped dye laser has been used to study photodetachment of NO_2^- in the photon energy range 2 to 3 eV. The photodetachment cross section at 2 eV is small ($\sim 2 \times 10^{-20} \text{ cm}^2$) but not zero. This result does not imply, however, that the electron affinity of NO_2 is less than 2 eV, for it presently appears that photodetachment at 2 eV is occurring from excited vibrational states of NO_2 . We can not at present identify the structure in the cross section, and thus are unable to resolve the question of the electron affinity of NO_2 .

The 35 kV nitrogen laser has now been completed and is fully operational. The output power is 1-2 MW in ~ 10 μ sec pulses, with a repetition rate of 10 pulses per second. The laser appears to be reliable, and its performance is as good as expected, based upon extrapolations from our smaller N_2 laser. A description of this work is being prepared for submission to the Review of Scientific Instruments.

The N_2 laser-pumped dye laser is being currently utilized in a series of molecular fluorescence studies in collaboration with Dr. H. M. Poland. We are studying intramolecular energy transfer in intermediate size molecules such as glyoxal. These experiments are presently in a preliminary stage, but results are expected shortly.

We have looked briefly at the photodetachment of CN^- using the N_2 laser directly. This experiment showed that the photodetachment threshold for CN^- is greater than 3.68 eV. While more detailed studies must await the development of a source of more energetic photons, it is clear that CN^- is among the most stable negative ions known, and may be a "terminal" negative ion in certain types of atmospheres.

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Molecular Dissociation Processes (Dr. L. J. Kieffer)

The major results of our recent work particularly the thesis research of Dr. R. J. Van Brunt were reported in two publications.^{1,2} It appears as though the angular distribution of molecular fragments yields an unambiguous assignment of the symmetry of the repulsive molecular state involved in dissociative electron attachment whereas for the dissociative ionization process this was not the case. The results of measure-

ments of the angular distribution of positive ion fragments from H_2 and D_2 showed that for these light molecules molecular recoil was very important. Whether the ambiguity of the results for H_2 and D_2 is due to some basic theoretical problem or some peculiarity in the structure of H_2^+ and D_2^+ is not clear.

We have recently turned our attention to the dissociative ionization process $e + O_2 \rightarrow O^+ + O + 2e$. Dr. George Lawrence, a JILA-LASP Visiting Fellow, participated in these experiments. The energy spectrum of the fast O^+ ions observed with our apparatus was compared with that observed by dissociative photoionization.³ The agreement was reasonable considering the energy resolution of the two experiments. This may imply that the same repulsive states of O_2^+ are excited both by electrons and photons. The symmetry of the repulsive O_2^+ states involved are not known. Observations of the angular distribution of the O^+ fragments may yield some information on the symmetries but our experience with H_2 and D_2 would imply that great caution must be observed in interpreting the results. Preliminary data indicate that it may be possible to observe the angular distribution of very low energy O^+ ions ($E < 1.0$ eV). These experiments are proceeding.

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Atomic Collisions Information Analysis Center (Dr. L. J. Kieffer)

A data bank of experimentally measured low energy electron collision cross sections exists and is being kept up to date. Cross section data for all atomic species and those molecules of interest to aeronomy,

astrophysics and plasma physics are included. Data are selected for the data bank on the basis of previous and current critical reviews of the appropriate measurement techniques.¹ Data are stored on magnetic tape and microfilm copies of graphical displays of all the data in the data bank are available in the Information Center. Graphical displays or data in tabular form are available on request. Compilations of these data are published in the open literature when appropriate.

A data bank of experimental photoabsorption and photoionization cross sections for all atomic species has the same status as the electron collision cross section data bank. Data are available on request. A bibliography on photoabsorption and photoionization data has been issued in report form.² In addition, a compilation of atomic data³ and critical review of molecular photoabsorption cross section data⁴ have been accepted for publication.

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III. Electron Energy Losses and Atomic Interaction Theory

Project Coordinator: Dr. E. C. Beaty

Electron Energy Losses (Dr. E. C. Beaty, Dr. C. Opal, Mr. W. K. Peterson)

The energy distribution and angular dependence of secondary electrons generated by the impact of electrons on He, Ne, Ar, Kr, Xe, H₂, N₂, O₂, NO, CO, H₂O, NH₃, CH₄, C₂H₂, and CO₂ have been measured over the 4 to 200 eV range. The measurements were made in a crossed-beam apparatus with the use of a fixed hemispherical electrostatic analyzer and a rotatable electron gun. The observed spectra were integrated over angle to obtain relative cross sections for secondary-electron production. It was found that the spectra of all the gases (except Ar, Kr, and Xe, which contain intense electron emission features in this energy range) could be described by the following simple function

$$\sigma(E_p, E_s) = \sigma(E_p, 0) / [1 + (\frac{E_s}{\bar{E}})^{2.1}] \quad ;$$

$\sigma(E_p, 0)$ is the cross section for an electron of primary energy E_p emitting an electron of zero energy. This can be related to the total ionization cross section providing a means of normalizing our data. \bar{E} is a characteristic energy which depends on the gas type but is independent of E_s and is only weakly dependent on E_p for He, O₂ and N. Although the spectra of the other gases have not been investigated at different primary energies, it seems likely that this single functional form would describe the secondary spectra for a particular gas at all primary energies greater than a few times the ionization potential. Table I shows how the characteristic energy, \bar{E} , depends on primary energy E_p for Ne, N₂ and O₂. Table II gives the values of \bar{E} for the other gases at 500 eV.

Table I. Dependence of shape parameter (\bar{E}) on primary energy (E_p).^a

Gas	E_p (eV)	E_{max} (eV)	\bar{E} (eV)
He	100	33	15.4
	200	67	15.5
	300	100	15.9
	500	167	16.4
	500	200	15.8
	1000	200	17.3
	2000	200	17.6
N ₂	100	33	12.7
	200	67	12.7
	300	100	13.3
	500	167	13.2
	500	200	13.0
	1000	200	14.3
	2000	200	14.1
O ₂	100	33	17.5
	200	67	17.6
	300	100	18.0
	500	167	18.1
	500	200	17.4
	1000	200	19.0
	2000	200	18.9

^aThe parameter was fit over the range of secondary energies between 4 eV and E_{max} .

Table II. Comparison of observed ejected electron spectrum shape parameter (\bar{E}) with corresponding ionization potential (I).^a

Gas	I	\bar{E}	\bar{E}/I
He	24.6	15.8	.64
Ne	21.6	24.2	1.12
Ar	15.7	(10.0)	(.64)
Kr	14.0	(9.6)	(.69)
Xe	12.1	(8.7)	(.72)
H ₂	15.4	8.3	.54
N ₂	15.6	13.0	.83
O ₂	12.2	17.4	1.43
CO	14.1	14.2	1.01
NO	9.5	13.6	1.43
H ₂ O	12.6	13.0	1.03
NH ₃	10.1	10.8	1.07
CH ₄	13.0	7.3	.56
C ₂ H ₂	11.6	10.0	.86
CO ₂	13.8	13.8	1.00

^aThe fits of our standard function to the Ar, Kr, and Xe spectra were poor; consequently, the values of \bar{E} are not very significant for these gases.

Atomic Interaction Theory (Dr. S. Geltman and Mr. M. B. Hidalgo)

Fine structure transitions in alkali-rare gas collisions. The intra-multiplet transitions in the first 2P state of alkali atoms induced by thermal collisions with rare gas atoms have been studied in a semi-classical theoretical treatment. The trajectories for the relative motion have been represented by hard-sphere collisions with the hard-sphere radius r_0 taken as an adjustable parameter to attempt to fit the precise experimental measurements of Gallagher^{1,2} for the depolarization ($j m_j = \frac{1}{2} \frac{1}{2} \rightarrow \frac{1}{2} - \frac{1}{2}$) and inelastic ($j = \frac{1}{2} \rightarrow \frac{3}{2}$) cross sections. Previous theoretical treatments³ have all used undistorted straight line trajectories and have yielded cross sections which were reasonable for the lighter alkalis but as much as 100 or 1000 too high for Rb and Cs.

The basis set of states was taken to be the six $j m_j$ states of the alkali doublet. The effective interaction potential (averaged over alkali radial coordinates), which contributes to the fine structure transition, has the asymptotic van der Waals form

$$\langle V \rangle \xrightarrow{\text{large } R} \frac{\alpha \langle r^2 \rangle}{R^6} Y_{20}^0$$

where α is the rare gas polarizability. We have evaluated higher terms in the multipole expansion (R^{-8} and R^{-10}) and found that, due to the multipole expansion being asymptotic rather than absolutely convergent, it was not practical to include them in our effective potential. Instead we joined the above van der Waals form to a short-range form given by the electrostatic field of the rare gas atom, as given by the Clementi tabulations⁴ of Hartree-Fock wave functions.

With the above constructed effective potential and model trajectories the 6 coupled equations are solved numerically to obtain the transition amplitudes. The result for the calculated depolarization and inelastic cross sections is given in Fig. 1 for Rb-Kr. Notice that there is a unique value of r_0 ($\approx 13.5 a_0$) for which both experimental cross sections can be obtained from our theoretical model. In the table below are the results we have obtained for Rb and He, Ne, and Kr. The simultaneous fitting of the depolarization and inelastic cross sections for the He and Ne cases is not as satisfactory as in the Kr case, but close enough to verify the essential correctness of our model for the process.

Collection of Results for Rb

Pair	r_0/a_0	$\sigma(1/2 \rightarrow 3/2)$		$\sigma(1/2 \ 1/2 \rightarrow 1/2 - 1/2)$	
		Expt/ \AA^2 [†]	Present/ \AA^2	Expt/ \AA^2 ^{††}	Present/ \AA^2
Rb-He [*]	6.9	.5	2	-	-
	7.6	-	-	9	9
Rb-Ne ^{**}	10.1	.07	.66	-	-
	10.3	-	-	6	6
Rb-Kr ^{**}	13.5	.7	.7	10.6	10.6

^{*} $v = 3 \times 10^5$ cm/sec

^{**} $v = 2 \times 10^5$ cm/sec

[†]Gallagher (Ref. 2)

^{††}Gallagher (Ref. 1)

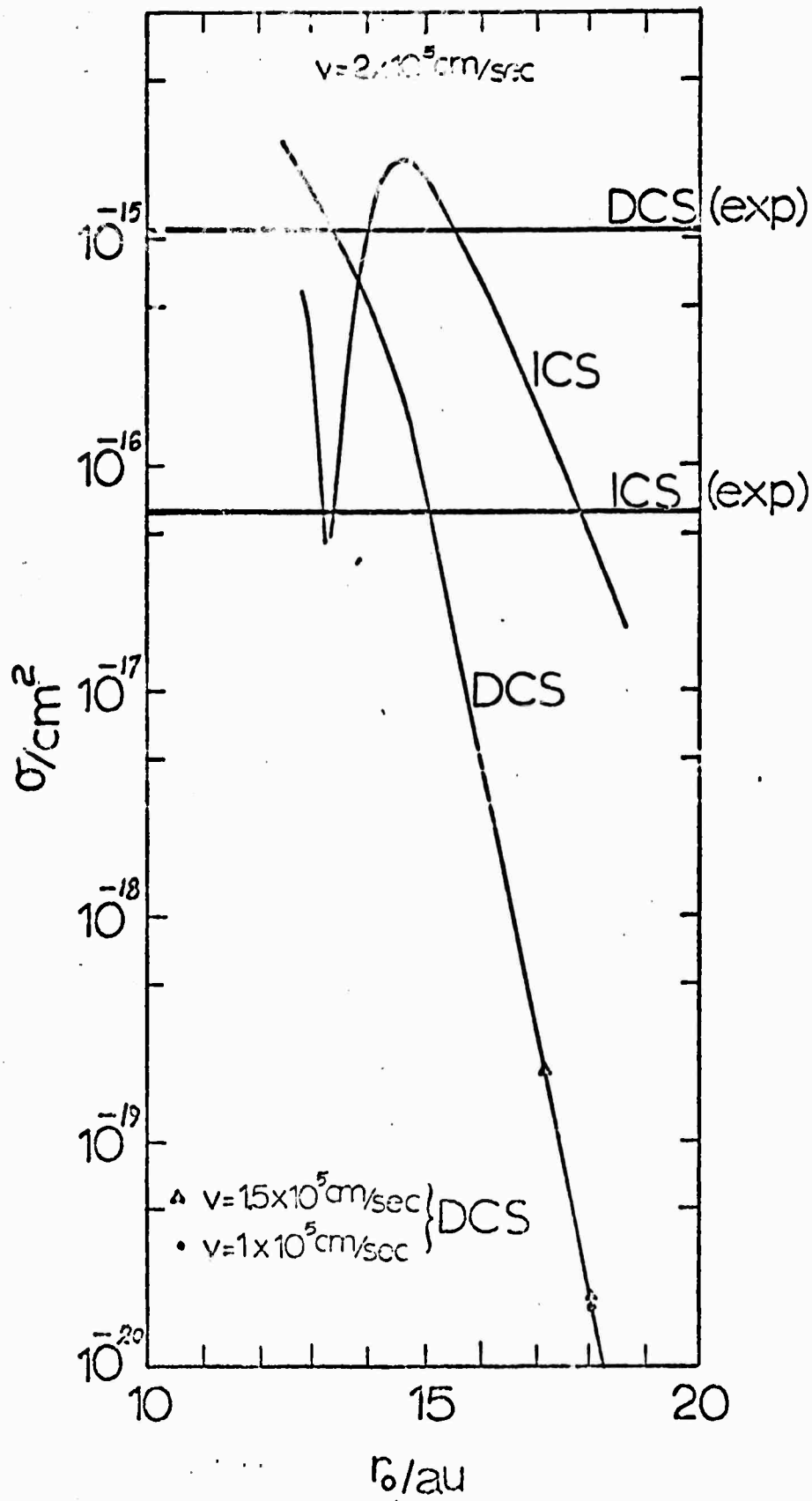


Figure 1

A Coulomb-projected Born approximation for high energy collisions.

In the description of a collision of a charged particle (p) with a hydrogen atom (e + n) the usual Born approximation for the T-matrix is of the form

$$T_B = \langle P_f | V_{pe} + V_{pn} | P_i \rangle ,$$

where $P_{i,f}$ represents an initial and final plane wave state. In the Coulomb-projected Born (CPB) approximation the V_{pn} interaction is included in the unperturbed part of the problem so that

$$T_{CPB} = \langle C_f | V_{pe} | P_i \rangle ,$$

where C_f is a Coulomb wave final state.

We have applied the CPB approximation to two cases of fundamental interest in atomic collision physics - (1) proton-hydrogen atom charge transfer and (2) electron impact hydrogen atom excitation. The charge transfer result has been compared in detail with two of the previous standard treatments, those of Brinkmann and Kramers⁵ and of Jackson and Schiff.⁶ We find the high energy limiting form $\sigma_{CPB} \xrightarrow{E \rightarrow \infty} .810 \sigma_{BK}$, as compared with $\sigma_{JS} \xrightarrow{E \rightarrow \infty} .661 \sigma_{BK}$. One advantage of our results over the Jackson-Schiff result is the absence of an anomalous back scattering peak. There is not enough data to establish the high energy form of the cross section experimentally.

In our second application of the CPB approximation we have evaluated the $1s \rightarrow 2s$, $1s \rightarrow 2p$, $1s \rightarrow 3s$ excitation cross sections of the hydrogen atom by electron impact. This application uses an attractive Coulomb wave while our first application used a repulsive Coulomb wave. In the Born approximation of this process the V_{pe} term in T_B has a vanishing contribution due to the orthogonality of the initial and final atomic states. However, in T_{CPB} the incident electron-proton interaction is

included in the Coulomb wave C_f . Our general result is that the CPB total cross section approaches the Born total cross section at high energies. However, we find significant differences in the differential cross section at large angles, and these differences persist to the high energy limit. This is illustrated in Fig. 2 where we compare results for the $n = 1 \rightarrow 2$ excitation of hydrogen with experimental values of Williams.⁷ Note that while the two theoretical approximations agree at small angles (so that total cross sections will coincide) there are order-of-magnitude differences at large angles, with the CPB result reasonably close to experiment. This casts doubt on the validity of the Born approximation for large angle differential cross sections even in the high energy limit.⁸

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I. Proton-Hydrogen Charge Transfer" and
"II. Hydrogen Atom Excitation by Electrons."

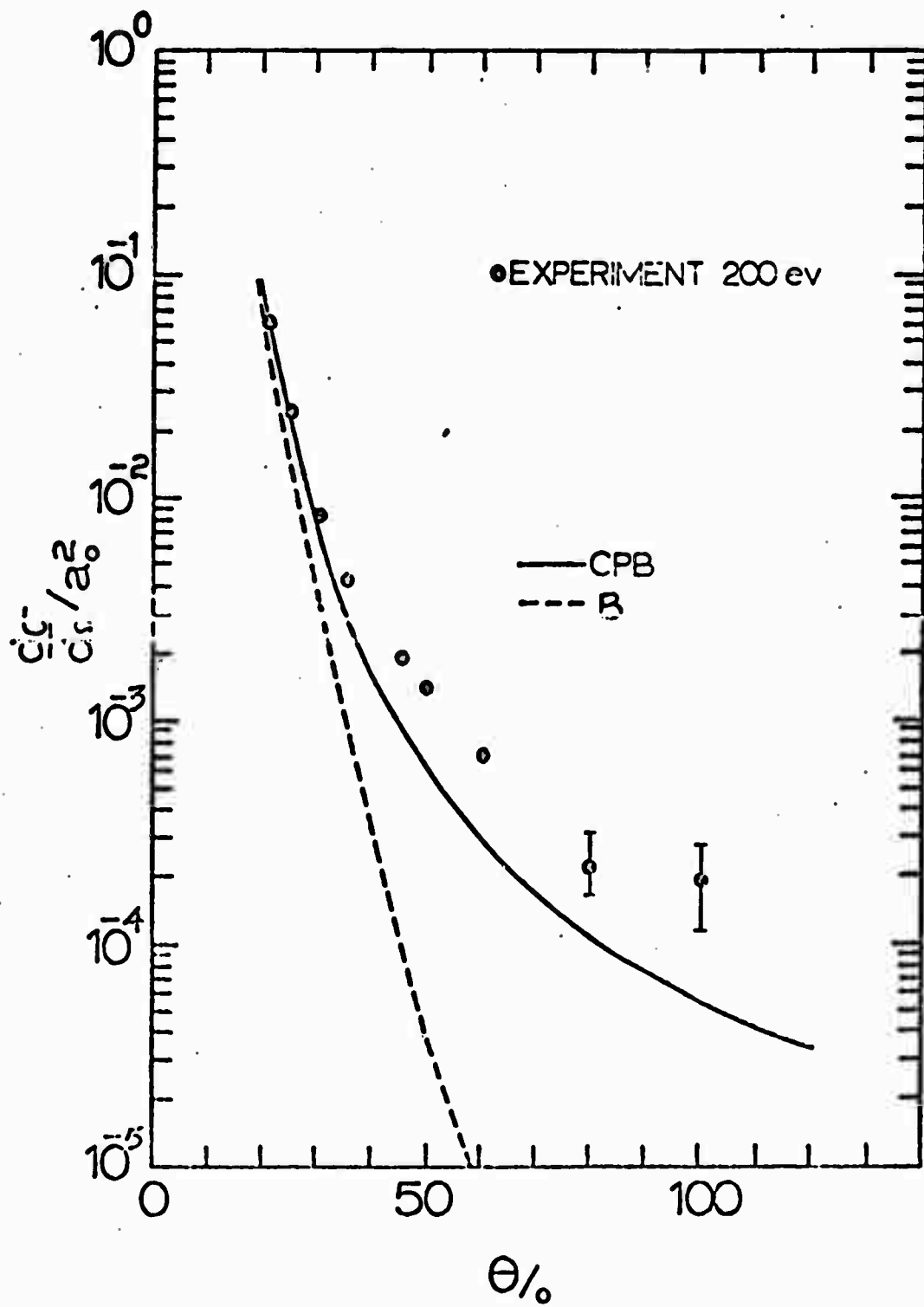


Figure 2

IV. Electron-Atom Collisions

Project Coordinator: Dr. S. J. Smith

Excitation of Ions by Electron Impact (Dr. G. H. Dunn, Dr. W. E. Kauppila and Mr. P. O. Taylor)

Measurements are being made in crossed beams of the cross sections for the processes:

- 1) $\text{Ca}^+(4S) + e \rightarrow \text{Ca}^+(4P) + e$
 $\text{Ca}^+(4S) \rightarrow \text{Ca}^+(4S) + h\nu(3965\text{\AA} \text{ and } 3934\text{\AA})$
- 2) $\text{Ba}^+(6S) + e \rightarrow \text{Ba}^+(6P) + e$
 $\text{Ba}^+(6P) \rightarrow \text{Ba}^+(6S) + h\nu(4934\text{\AA} \text{ and } 4554\text{\AA})$
- 3) $\text{N}_2^+(\text{X}^2\Sigma_g^+) + e \rightarrow \text{N}_2^+(\text{B}^2\Sigma_u^+) + e$
 $\text{N}_2^+(\text{B}^2\Sigma_u^+) \rightarrow \text{N}_2^+(\text{X}^2\Sigma_g^+) + h\nu(3914\text{\AA})$

Polarization of the resultant radiation is also being measured.

Beams of Ca^+ and Ba^+ are formed by surface ionization of the respective atoms, thereby precluding the metastable D levels from the beam. Ions of N_2^+ are formed by electron bombardment of N_2 . The ion beam of about 500 eV energy is crossed at right angles with a magnetically confined electron beam and the resultant radiation is observed along the third orthogonal axis through a lens-interference filter-photomultiplier system. Chopping of the two beams and appropriate gating of the two scalars used allows one to separate signal from background and to identify and eliminate spurious effects. Cross sections are obtained from the beam magnitudes, their spatial overlap, and the observed signal. Polarization is measured using a polaroid with its axis placed alternately parallel and perpendicular to the electron beam axis.

Preliminary measurements have been made with Ca^+ and N_2^+ (reactions

1 and 3). Figure 3 shows polarization measurements for the H and K lines of Ca^+ and also shows calculations of Saraph¹ using close-coupling matrix elements of Burke and Moores.² The agreement of experiment and theory is considered good. Figure 4 shows the cross section for excitation of the H and K lines as a function of electron energy normalized to Burke and Moores² calculation at their highest energy. (Such normalization is considered as a reasonable interim procedure on the basis of the good agreement between measured polarization and the close-coupling related calculated polarization.)

Also shown in the figure are unpublished distorted wave calculations made by Alan Burgess in this institute recently. A semiclassical estimate by Burgess of cascade effects is also shown.

Figure 5 shows preliminary results for 3914Å excitation of N_2^+ . These measurements differ by up to a factor of 25 with the results of Lee and Carleton.³ It is clearly important to get better measurements, since if Lee and Carleton's values are correct, this is an important electron energy loss process in disturbed atmospheres.

Presently preparations are being made to make the measurements on an absolute basis, so that no recourse need be made to normalization. For this, a moveable point source has been built and is being compared with a copper-point black body.⁴ It will be used to map out the absolute photon sensitivity of the apparatus as a function of position, which will then have to be folded with the beam current distributions to obtain cross sections. Development of these procedures is proceeding satisfactorily.

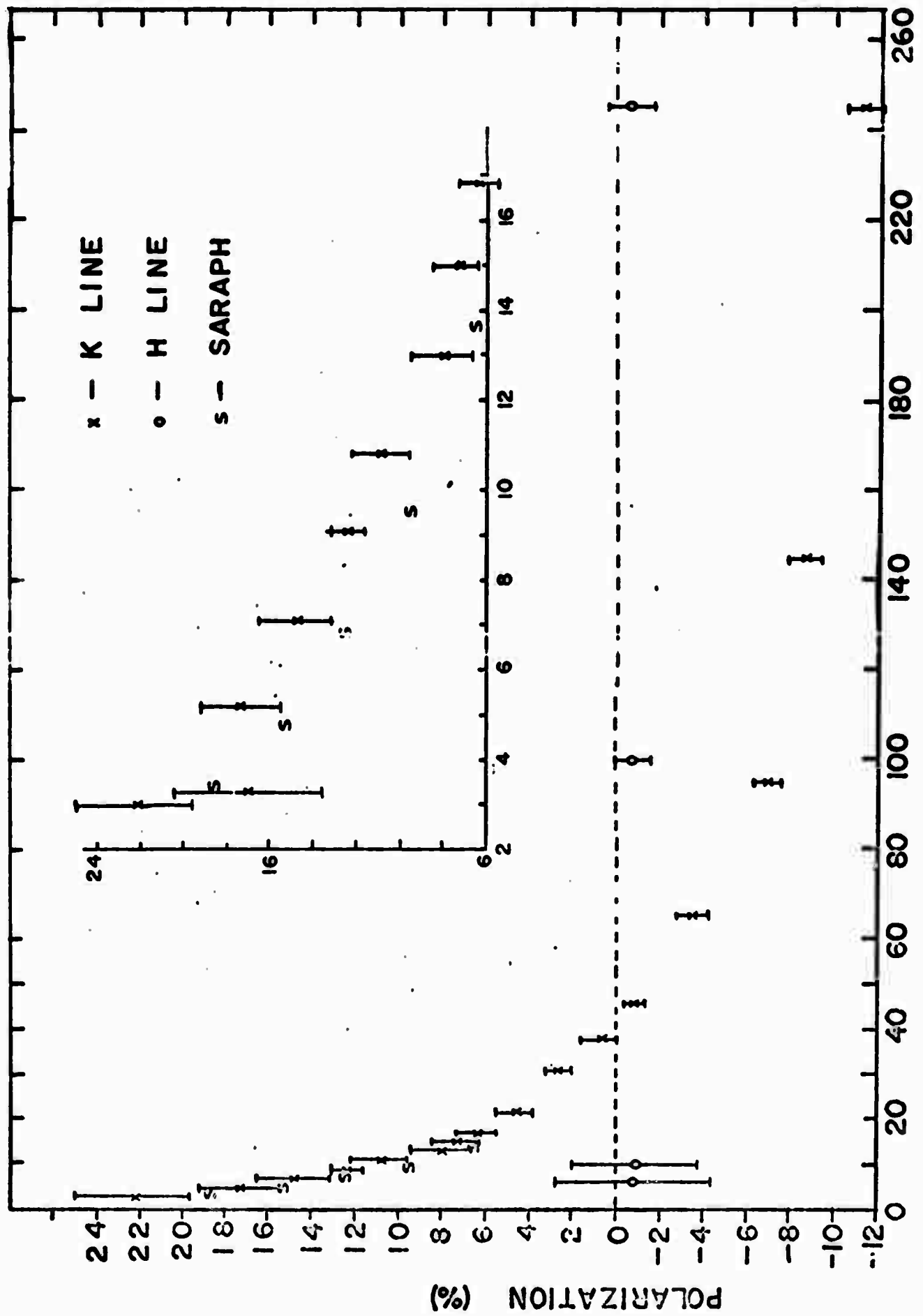


Figure 3

ELECTRON ENERGY (eV)

POLARIZATION (%)

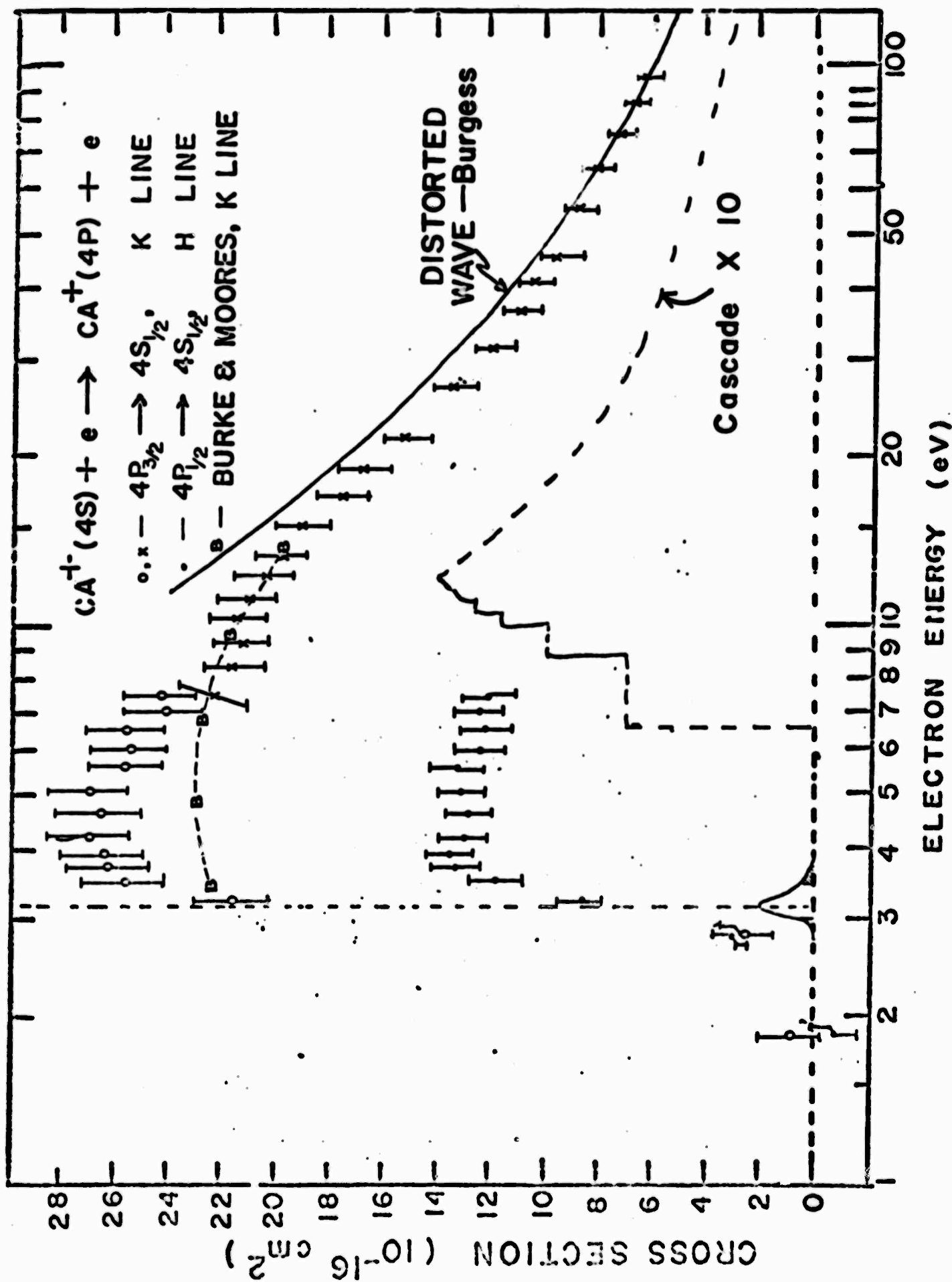


Figure 4

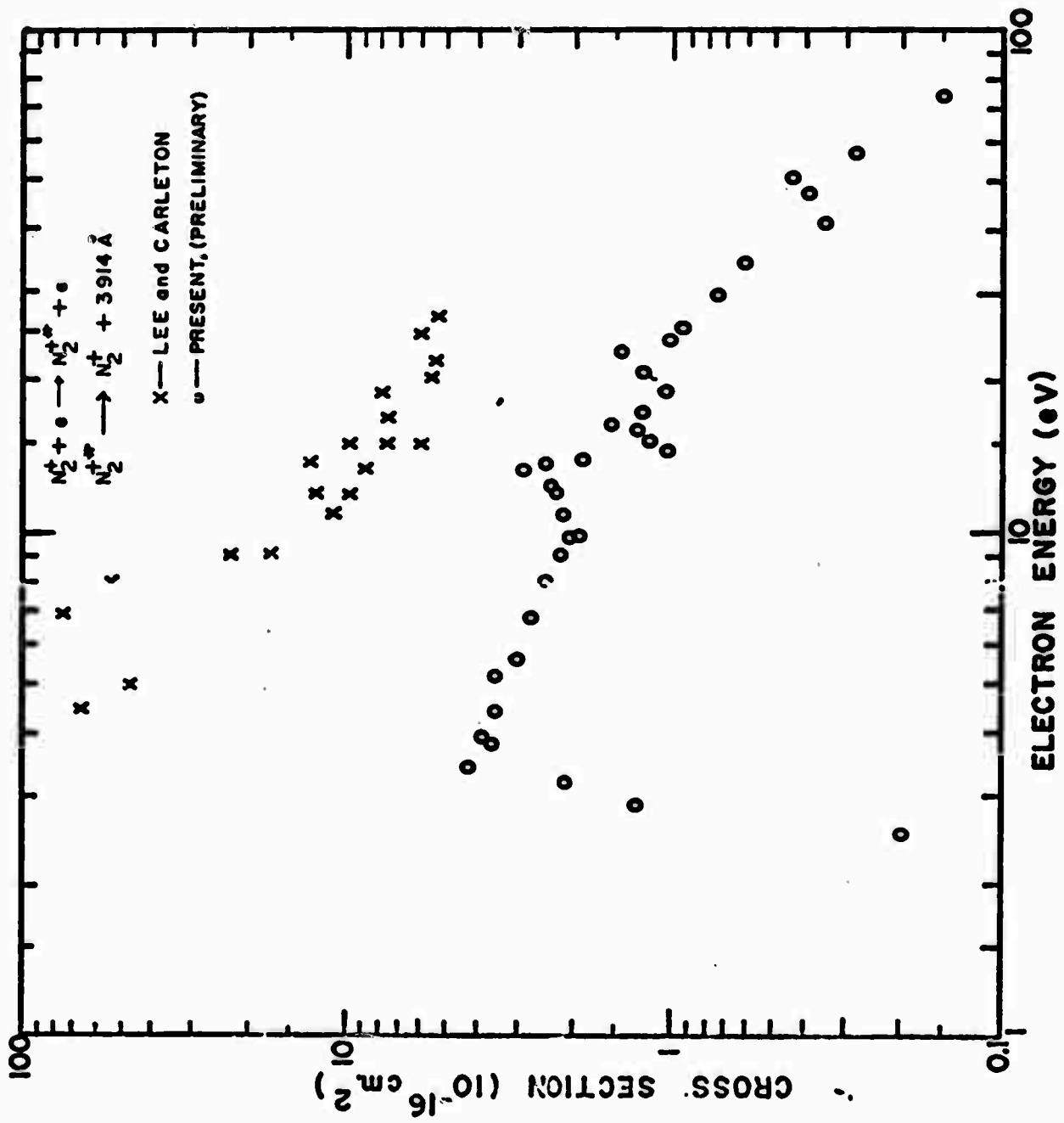


Figure 5

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4. R. D. Lee, NBS Technical Note 483, May 1969.

Electron-Ion Recombination (Dr. F. L. Walls, and Dr. G. H. Dunn)

A new technique is being developed for the study of electron-ion collisions. First studies are of the dissociative recombination of electrons and N_2^+ .

The N_2^+ ions will be formed and contained in an axially symmetric quadrupole ion trap of the Penning variety.¹ The stored gas of N_2^+ will be irradiated with a beam of electrons and the number of remaining ions vs time measured. The absolute number of ions, their spatial distribution, the electron beam intensity, and its spatial distribution will be measured using a Bendix Spiraltron Bundle Detector. These measurements are sufficient to determine the cross section for a given electron energy and distribution of ion vibrational levels.

The apparent advantages of this approach over earlier ones are: The translational motion of the stored ions can be coupled to a suitably tuned LC circuit allowing one to non-destructively monitor the number and the translational temperature of the stored ions.² This process, coupled with ion-ion collisions, also causes the translational motion and the occupation numbers for the vibrational levels of N_2^+ to assume a Boltzmann distribution corresponding to the noise temperature of the LC circuit. The noise temperature can be easily manipulated either electronically or with an external heat bath. This allows one to separately investigate the effects of the interaction energy (which is primarily determined by the electron energy) and the distribu-

tion of occupied vibrational levels, on the cross section.

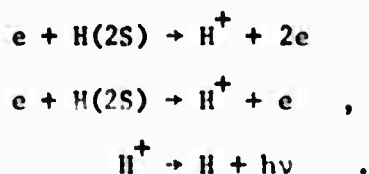
An ion trap and vacuum system have been built. The vacuum system has a base pressure of approximately 3×10^{-11} torr equivalent N_2 pressure as measured on a GE Triggered Discharge Gauge. Vacuums of this order of magnitude are required to minimize ion losses from ion-residual gas collisions. The electron gun functions according to design theory³ and delivers about 5×10^{-11} amps at 100 mV FWHM. Straightforward revisions are being made which should give greater than 10^{-10} amps at 25 mV FWHM.

The next step is to investigate the storage and manipulation of N_2^+ gas in the present trap.

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Electron Collisions with Metastable Atoms (Dr. G. H. Dunn and Dr. W. E. Kauppila)

Preparations have been made to study the processes:



The experiment is as follows. A beam of protons from a duo-plasmatron source charge transfers in a cesium cell to give metastable atoms. The metastable atom beam intensity is measured by photoionization with an N_2 laser. The known metastable atom beam is crossed with a variable energy electron beam and resultant protons and photons are detected and measured. Relevant cross

sections can then be computed.

The ion source is built and tested, the N_2 laser is built and tested, and the charge transfer chamber has been built. These components must be used with the ion beam machine being used for excitation studies discussed in the previous section, and further progress with the experiment awaits termination of that work.

Electron Impact Excitation of Hydrogen (Dr. S. J. Smith and Mr. A. H. Mahan)

This electron-hydrogen atom crossed beam experiment utilizes a solar-blind photomultiplier with an oxygen window to detect Lyman- α photons emitted from the interaction of the crossed beams. If the interaction region is field-free (except for small space charge fields and residuals of the shielded magnetic field of the earth) the Lyman- α observed is attributed to radiative decay from the short-lived $2p$ state of atomic hydrogen. In this work we have introduced a radio-frequency field tuned to the $2P_{1/2} - 2S_{1/2}$ interval (Lamb shift) to mix the two states and provide a mechanism for quenching the ($2S_{1/2}$) metastables. With the rf field turned on, the observed Lyman- α flux contains components due to excitation, both direct and by cascades, into the $2S_{1/2}$ state. With appropriate attention to the quenching efficiency, the ratio of signals with rf on and rf off can be used to obtain the ratio of the $2s$ to $2p$ excitation cross sections. In these measurements the mass 2 hydrogen isotope (deuterium) was used to simplify the quenching problem which is dependent on hyperfine structure.

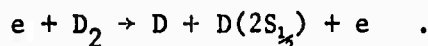
This ratio was studied at energies ranging from threshold to 500 eV. By synchronizing the scaler gates with a rotating atom beam chopper photons were counted in two channels, representing background gas excitation and background plus beam excitation. In order to obtain the $2s/2p$ ratio with statistical accuracy of 5 to 10%, approximately 10^5 counts (requiring approxi-

mately an hour of data collection) were necessary at each energy.

As a by-product of this work an excitation cross section for $H(2p)$ is obtained with a statistical accuracy of a fraction of a per cent. Since these measurements were carried out to 500 eV (our previous 2p measurements extended only to 200 eV) we expect them to provide a very valuable check on the validity of the Born approximation and other theoretical methods.

We are presently preparing these results for publication, probably in The Physical Review. The work will also be discussed in a contributed paper at the Washington meeting of the American Physical Society. Figures 6 and 7 show these 2p and 2s excitation results in comparison with recent pseudo-state calculations by Burke and Webb,¹ and with experimental results of McGowan et al.² (for 2p) and Kauppila et al.³ (for 2s). Our 2s cross section is scaled through our measurement of the $\sigma(2s)/\sigma(2p)$ ratio to a normalization to the Born approximation for the 2p excitation cross section; the ratio is on the basis of a preliminary evaluation of the quenching efficiency.

The rf quenching technique described above was also used for an absolute measurement of the cross section for the dissociative excitation process



This measurement was accomplished by filling the vacuum apparatus with a static pressure of deuterium gas. Careful measurement of the gas pressure using a McLeod gauge (exhaustively studied and corrected for secondary effects), and absolute calibration of the Lyman- α detector lead to a value for the cross section of 3.3×10^{-18} ($\pm 20\%$) cm^2 at 39 eV. The relative cross section was also measured. This work was carried out by Mr. Donald Cox, and is described in full in his Ph. D. thesis.⁴ This work is also being prepared for publication in The Physical Review.

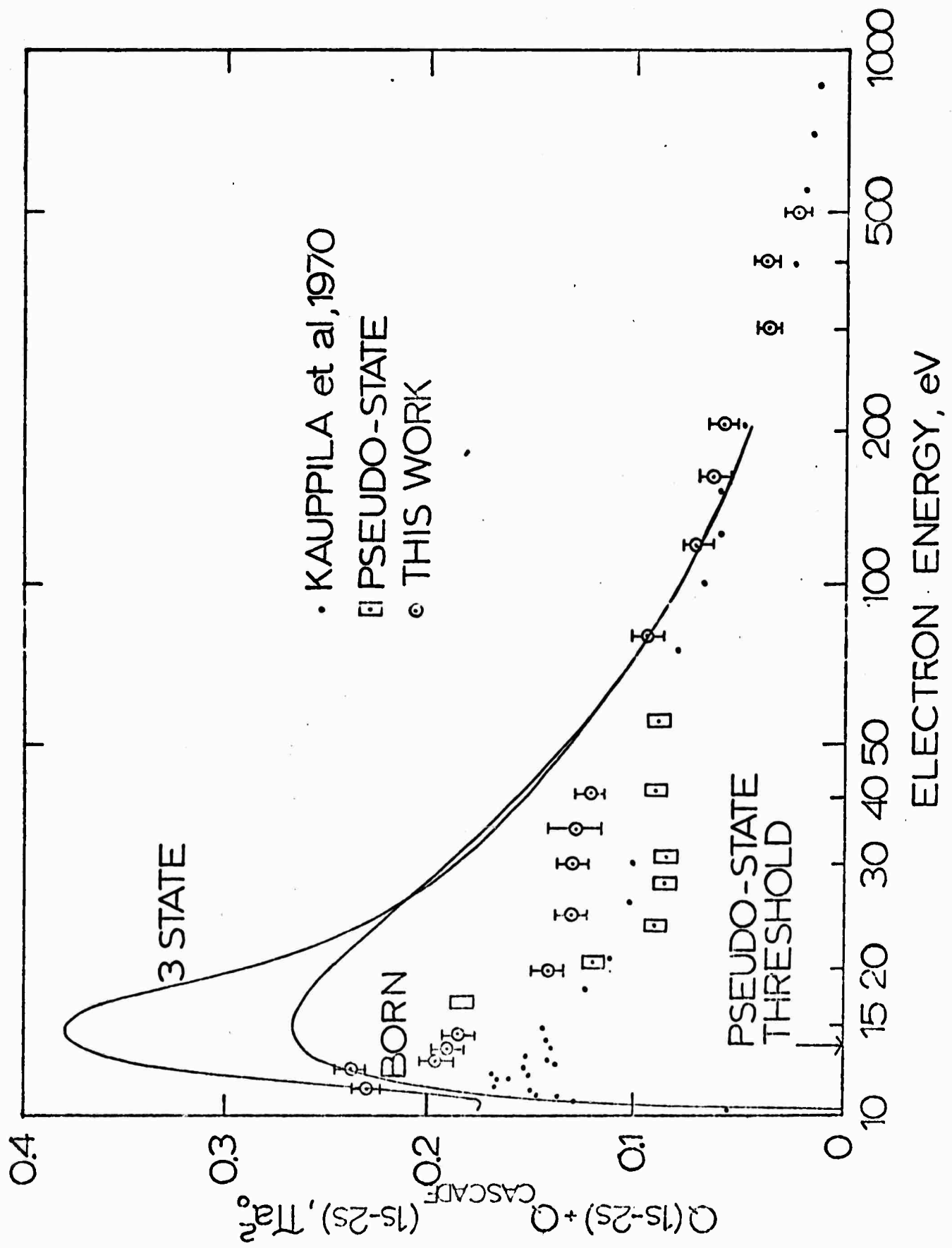


Figure 6

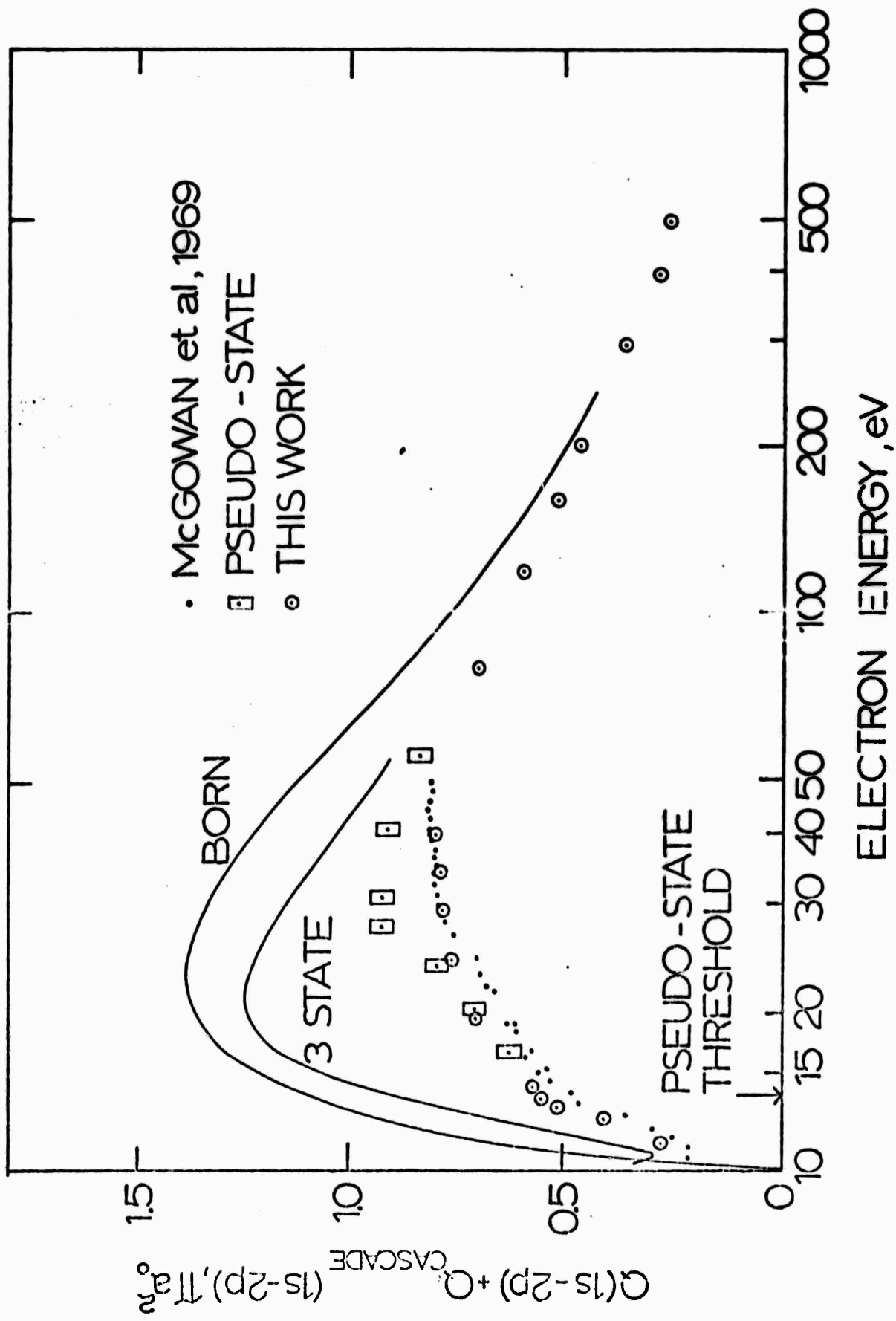


Figure 7

On completion of the Lyman- α work the apparatus was modified for a rather sophisticated study of the Balmer radiation excitation by electron impact. Fast modulation of the electron beam is to be used to allow time resolution of components of the Balmer radiation arising in states of differing mean lifetimes. This will allow separation of cascade components and may allow resolution of the Balmer- α components arising from s, p and d states of the $n = 3$ shell. This work is being conducted as thesis research by Mr. Harvin Mahan. Background light levels have been reduced to permit dc measurements of Balmer- α excitation.

In a preliminary measurement with a dc electron beam we measured the cross section for excitation of the Balmer- α line at energies ranging from 15 to 500 eV. The atomic hydrogen beam was produced by thermal dissociation of molecular hydrogen at approximately 2500°K; the electron source was a Soa type gun of half-width 0.35 eV. Observations were made at 90° to the electron beam axis. The present data are not corrected for cascading or polarization. The results are higher than those of Kleinpoppen and Kraiss⁵ at energies below 50 eV, but agree well at higher energies. Our results also are in fair agreement with the calculations of Morrison and Rudge.⁶

Electron-Alkali Atom Scattering (Dr. S. J. Smith, Dr. M. V. McCusker, and Mr. D. Hils)

Toward the end of the period covered by this report the first extensive data runs were carried out in a measurement of the polarization of electrons scattered from a polarized alkali atom beam. The apparatus is shown schematically in Fig. 8. The atom beam, presently potassium, is formed by thermal effusion through an orifice. It is chopped and then passed through a hexapole magnet which throws out one component of the polarization and focuses

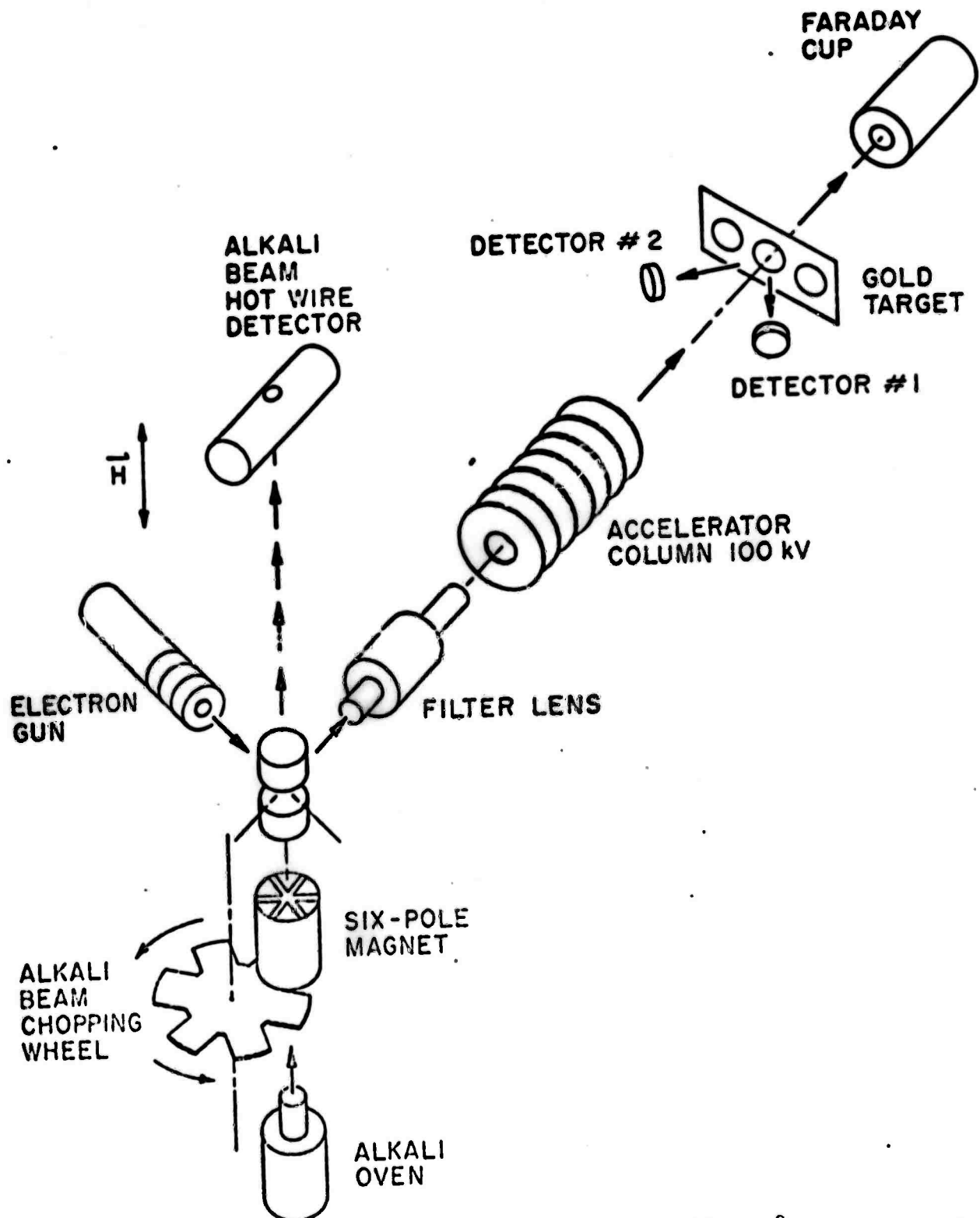


Figure 8

the other. In the region beyond the hexapole magnet a very weak but well-defined magnetic field, as low as 10 mG, maintains the directional polarization along the axis of the atom beam (but at a decreased value of polarization because of hyperfine coupling) throughout the region of interaction with the electron beam. The electron gun, mounted on a rotating table, operates transversely to the polarization axis. Electrons scattered into a selected angle in the plane perpendicular to the atom beam axis are collected, run through a filter lens to remove inelastically scattered electrons, and accelerated to 10 kV. Two surface barrier lithium drifted detectors are used to measure the asymmetry of scattering from a gold foil (Mott effect). Pulses are light piped out of the high voltage region and sent into four scaler channels, two for each detector being synchronized to the chopping of the atom beam.

First data are being taken at 40° , and at about 4 eV electron energy. The objective is to provide a test for recent close coupling calculations by Burke⁷ and Karule,⁸ which predict strong angular dependences and energy "resonances" in the polarization. The data should therefore provide a sensitive test of the theory. As of this date we have made a preliminary measurement of electron polarization at one angle (40°) and one energy (3.5 eV). A value of $|f|^2/o = 0.85 \pm 0.1$ has been computed from this measurement.

We also plan to make careful measurements of the angular distribution for elastic scattering. The ease and accuracy with which this is done is illustrated in the data shown in Fig. 9. We have made measurements at several scattering angles between 15° and 87° ; the counting time was determined by observing the accumulation of charge from the alkali beam detector on an electrometer and turning off the scalers manually after the charge reached an arbitrary level. No compensation or manipulation of the data of any sort was done. Despite the crudeness of these methods the data show remarkable

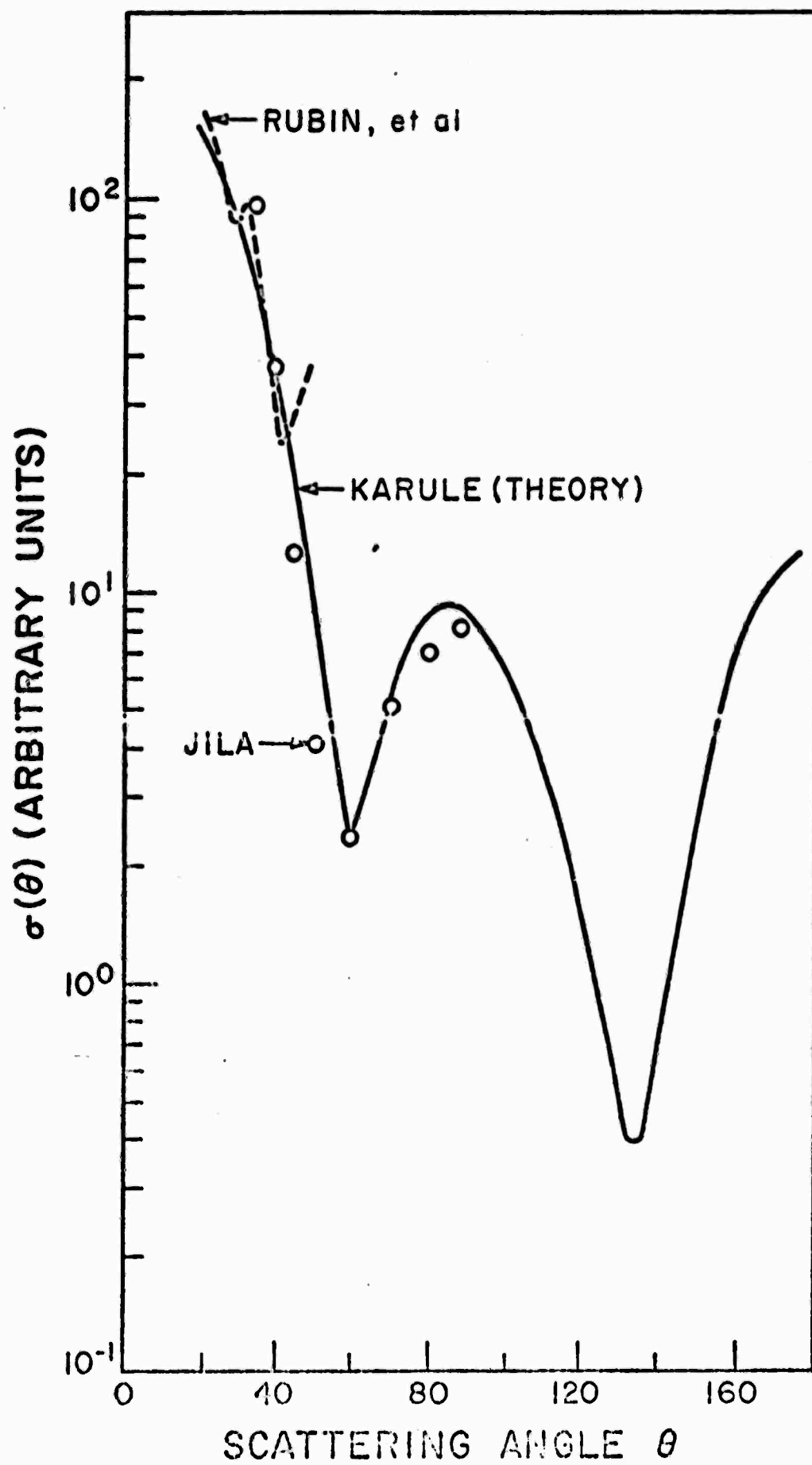


Figure 9

agreement with the theoretical computation of Karule (also shown in the figure). Our data were normalized to Karule's curve at $\theta = 60^\circ$.

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4. D. Cox, Ph.D. thesis, University of Colorado, 1970 (unpublished).
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Collisional Relaxation Mechanisms (Dr. P. L. Bender and Mr. R. Brill)

Work has been going on for some time to try to obtain nuclear orientation in the ground state of Xe^{129} by optical pumping with the 1470\AA resonance line. The method used involved circular polarization of the light by Brewster's angle reflection from a LiF plate followed by passage through a stressed LiF quarter-wave plate. MgF or LiF windows were used on the lamp and sample, and the entire optical path had to be in vacuum to prevent absorption of the light by air.

The purpose of the work has been to try to study relaxation of the Xe^{129} nuclear magnetic moment by collisions with the walls of the sample. Each time that the atom hits the walls it sticks for a short time before re-evaporating, and some relaxation occurs during the sticking time. Relaxation times of up to 100 sec had been obtained at the Ecole Normale

Superieure in Paris for Hg atoms in heated quartz samples. It was expected that the more inert Xe atoms would stick to the walls for a shorter time and give a longer relaxation time.

The measurement of long relaxation times in Xe would open up the possibility of an improved spin-precession gyroscope. Such gyroscopes require the accurate measurement of precession frequencies for two different isotopes with different nuclear moments. From the two different frequencies one can determine both the magnetic moment at the sample and the rate of rotation of the apparatus with respect to inertial space. He^3 has a 10^6 sec or greater relaxation time, but the accuracy of the device would be limited by the relatively short relaxation time of the Hg or other second species which was used. Obtaining long relaxation times with Xe would also permit a more accurate check on the isotropy of inertial properties of matter.

Unfortunately, the direct approach to optically pumping Xe^{129} has not been successful. Apparent signals indicating orientation were obtained, but it was not possible to reproduce them later. A number of improvements in the apparatus were made which decreased the noise level, but no signals were obtained after this. Checks were also made to see that the various parts of the apparatus were operating as expected. It thus appears that the xenon atoms may be getting relaxed considerably more rapidly than expected. Because of the ultraviolet transmission requirement for the sample cell, it was unfortunately not possible to use a cell made simply of pyrex or quartz. A pyrex cell with LiF windows attached by epoxy was used, and relaxation on the epoxy or LiF may have reduced the relaxation time to below the observable level.

In view of the difficulties experienced in obtaining any signal at all by conventional optical pumping, the prospect for using Xe in a gyroscope or for the isotropy of inertia experiment seem small. However, before stopping work on the project it seemed worthwhile to try one alternate approach to see whether a short relaxation time was really the problem. In a fairly strong magnetic field, Lehmann¹ has shown that inequalities in hyperfine coupling can lead to an asymmetry in the pumping by right and left circularly polarized light. For Xe¹²⁹ the pumping is enough more efficient at 400 G so that a net 20% pumping efficiency with unpolarized light should be achieved. The high field method was not tried earlier because it would not be attractive for use in the desired applications.

The high field method has several experimental advantages. Most important, the removal of the space-consuming and inefficient polarization optics allows the lamp to be moved up close to the cell, thereby effecting, roughly, a three order-of-magnitude increase in light flux increase in the pumping rate, plus the lower relative shot noise level, should make it possible to see optical pumping effects despite relatively short relaxation times. Also, the difficult quarter-wave plate adjustment is eliminated.

Preliminary attempts were made during the period covered by this report to produce an orientation by optical pumping in a 40 G magnetic field. This would give a theoretical pumping efficiency of about 4% for equal intensities of the $F = 3/2$ and $1/2$ hyperfine components of the resonance line. A portion of the transmitted light through the cell was observed with a solar-blind photomultiplier. An rf field at the ground-state Zeeman frequency was switched on and off to destroy the orientation,

and a signal at the switching frequency was looked for with a synchronous detector. No signal was observed under conditions where a change of a few parts in 10,000 in the transmitted light would have been detectable.

The interpretation of transmitted light experiments is now being investigated. The orientation produced in the sample and the sensitivity of the transmitted light to the orientation need to be calculated for the actual case where the hyperfine component intensities are different. The results must then be integrated over the length of the sample. If the calculations indicate that transmitted light measurements have good sensitivity, the experiment may be repeated in a higher magnetic field. Otherwise, a decision will have to be made on whether it is worthwhile to change the experimental setup so that scattered light can be observed.

1. J. C. Lehmann, J. de Phys. 25, 800 (1964).

V. Radiation Processes

Project Coordinator: Dr. A. C. Gallagher

Collisional Radiation (Dr. A. C. Gallagher)

Measurements of the far wing emission spectrum of Cs resonance lines broadened by inert gas collisions have been completed. Wing intensities from 100 to 1000 cm^{-1} from line center are measured relative to the intensity at the line center for optically thin Cs excited by D_1 and D_2 lines from a resonance lamp in the presence of 100 - 400 torr inert gas over a temperature range of 300 - 800°K. The data are being analyzed in terms of the quasi-static theory of line broadening. Provided there is a single valued relationship between the potentials for the excited states ($A^2\Pi, B^2\Sigma$) of the cesium-rare gas molecule and the differences between these potentials and the ground state potential ($X^2\Sigma$), the temperature dependence of the wing intensities can be related to the distribution in energy and internuclear separation of atoms in the excited state. Such a uniqueness is predicted by Baylis's potentials,¹ so that it is possible to derive the A, B, and X potentials from these experimental results. Differences in the distribution functions in energy and internuclear separation for binary collisions and for bound molecules indicate that at the pressure of these experiments the vibrational populations of the $(\text{Cs Xe})^*$ and $(\text{Cs Kr})^*$ reach equilibrium during the radiative lifetime. Analysis of these data will continue during the next report period. In addition, we will begin measurements of the far wing emission of rubidium resonance lines broadened by rare gases. Rubidium is of particular interest because the larger signal-to-noise ratio available using recently available photomultipliers will allow measurements at lower rare gas densities where relaxation effects among the vibra-

tional states of the excited rubidium rare gas molecule may become observable.

Electron Excitation of Metal Atoms (Dr. A. C. Gallagher and Mr. E. Enemark). Measurement of the energy dependence of the polarization and the apparent electron impact excitation function for the sodium 3s - 3p transition has been completed using a crossed beam apparatus. The incident electron energy range extended from threshold (2.1 eV) to 1000 eV and the electron energy spread was about 0.3 eV. The polarization of the unresolved doublet radiation (5890 - 96Å) was measured to an accuracy of a few percent. The measured polarization is in agreement with the logarithmic approach to the polarization limit at infinite energy as calculated by Percival and Seaton.² Our results also agree with the calculated polarization value of Karule³ near threshold. After applying polarization and 5-6% cascade corrections, the excitation cross section for the 3p state was found to vary with energy at energies above 250 eV as expected for an optically allowed transition with an oscillator strength of 0.9. By normalizing to the first Born approximation at energies above 250 eV, fairly good agreement is found with the low energy cross sections of Karule and Peterkop.⁴ The cross section passes through a broad maximum of $43 \times 10^{-16} \text{ cm}^2$ at energies between 7 and 10 eV.

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Negative Ion Studies (Dr. J. L. Hall, Dr. R. J. Celotta, and Mr. R. Bennett)

In order to fully establish the initial and final vibrational states of the photodetachment transitions we have observed in NO^- and O_2^- we have used three techniques. The first was to measure the anharmonicities in the observed energy levels and correlate these with the neutral, spectroscopically derived levels. This measurement required an analysis of the small systematic effects present in the electron monochrometer and has been described in previous reports. The second technique involves the use of computer generated Franck-Condon factors which link the negative ion and neutral wave functions. It is possible to arrive at transition state assignments by observing that the relative intensities of neighboring transitions would be best explained by a particular choice of state assignment for any realistic negative ion potential. Some refinements of this technique have been made during the past six months but the most progress has been made on the third independent method of transition determination, that of isotope shifts. By using isotopes of the negative ion with the same electronic structure but different reduced masses it is possible to change the vibrational energy levels in an easily calculable way. By measuring the change in outgoing electron energy from a single transition for two different isotopes, it is possible to determine the transition state assignments. To accomplish this experimentally, a programmer was constructed which switches the ion mass under study and the monochrometer transmission energy every 8.3 msec. The resulting photodetachment spectra are digitally accumulated and the measurement of the small isotope shifts can be accomplished. During the past six months the programmer has been used to make precise measurements of the isotope shifts for two transitions in NO^-

and two in O_2^- . These measurements confirmed our previous state assignment.

Additional experimental work was performed to measure the angular distributions of outgoing photoelectrons. The expected angular distribution is of the form $1 + \beta P_2(\cos \theta)$ where P_2 is the Legendre polynomial and θ is the angle between the electric field vector of the light and the electron collection direction. The asymmetry parameter, β , can take on values from +2 (a pure $\sin^2 \theta$ distribution) to -1 (a pure $\cos^2 \theta$ distribution). By measuring the value of β for each final vibrational state we obtain the variation of β as a function of outgoing electron energy. For the NO^- and O_2^- ions β tends to be near -1 for photon energies a volt or so above threshold. We have measured the angular distributions corresponding to each transition in O_2^- during the past six months.

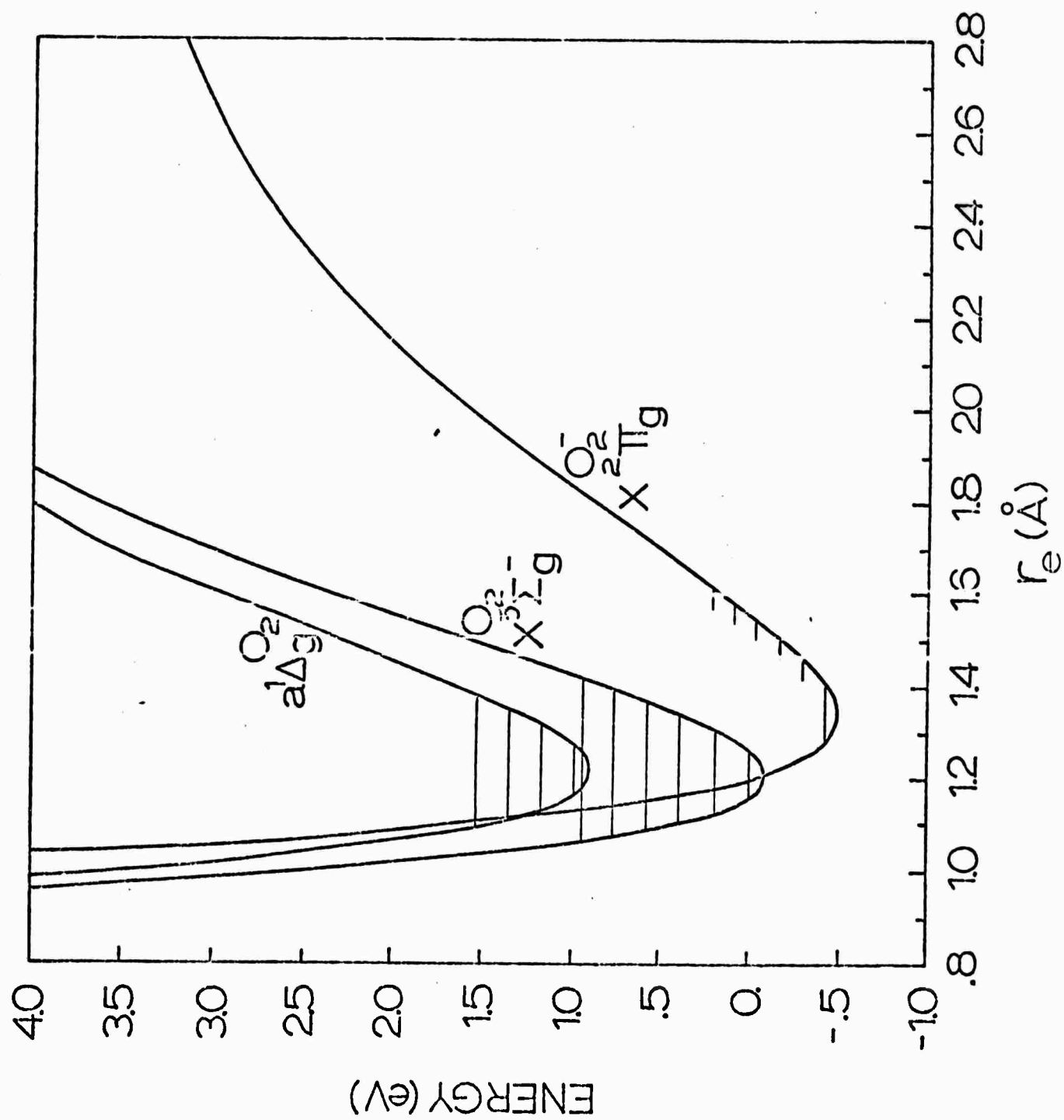
Additional work has been done to calculate the final corrections required to produce electron affinities for O_2 and NO . While very small corrections are still to be finalized, the present values and their error estimates are,

$$\begin{array}{ll} EA(O_2) = 0.425 \pm .030 \text{ eV} & EA(NO) = .022 \pm .020 \text{ eV} \\ \omega_e(O_2^-) = 1089 \text{ cm}^{-1} & \omega_e(NO^-) = 1472 \text{ cm}^{-1} \\ r_e(O_2^-) = 1.341 \pm .010 \text{ \AA} & r_e(NO^-) = 1.258 \pm .010 \end{array}$$

Figure 10 shows the O_2^- internuclear potential we have derived from our experiments. The energy levels shown as solid lines are the initial and final states of our observed transitions.

We are in the process of preparing a rather lengthy paper on our photo-detachment technique outlining in great detail the finer points of the procedure and the data analysis. In addition, we have been looking at the

negative ions, NH_2^- , S^- , SO^- , and SO_2^- . We find that the reported electron affinity for NH_2 , 1.2eV, is most probably wrong and that the true value seems to be $.770 \pm .05\text{eV}$. The SO^- ion will most likely be the next system we study and we shall then try to extend our analysis technique to triatomics and study SO_2^- .



Oscillator Strengths and Transition Probabilities (Dr. R. H. Garstang)

Calculations have been continued on transition probabilities of forbidden lines of a number of atoms and ions of particular interest. One atom studied, which had never been studied previously for this purpose, was Fe I.¹ Calculations were desired in part because the electron configurations involved were $3d^6 4s^2$ and $3d^7 4s$, and no studies had been made of forbidden atomic transitions in any atom or ion involving these two configurations. The results would also find application to the problem of the solar iron abundance. The interaction between the $3d^6 4s^2$ and $3d^7 4s$ configurations was included, as well as the usual spin-orbit interactions. When the transition probabilities had been obtained they were used to predict the equivalent widths of [Fe I] lines in the solar sunspot spectrum. A search for the strongest lines was made on spectra taken especially for this purpose at the Kitt Peak National Observatory. Two of the lines were possibly detected, the others being blended or otherwise not observable. The strengths of these two seem to confirm the high solar abundance of iron which had been previously found on the basis of Garstang's² transition probabilities for [Fe II] and on the basis of the Kiel f-values for Fe I and Fe II. Apart from its astronomical significance, we note the wide implication of these results, that there appear to be no gross errors in the absolute scales of the transition probabilities of forbidden lines. While checks of their absolute values were previously available³ only for [O I] and for certain heavy elements (e.g. [I I]) the additional check for an atom in the middle of the Periodic Table is a valuable one to have done, and increases our confidence in the calculated values for any element.

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3. R. H. Garstang, Mém. Soc. Roy. Sci. Liège, Ser. V, 17, 35 (1969).

Manuscripts Involving ARPA Funds

Listed below are papers submitted for publication during the period covered by this report. Co-authors not connected with JILA are shown in parentheses.

- T. F. Adams, G. B. Rybicki, and D. G. Hummer, "Numerical evaluation of the redistribution function $F_{II-A}(x, x')$ and of the associated scattering integral," J. Quant. Spectrosc. Radiat. Transfer, to appear.
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